

Electron localization in self-assembled Si/Ge(111) quantum dots

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Electron localization in the Si/Ge heterosystem with Si quantum dots (QDs) was studied by transport and electron spin resonance (ESR) measurements. For Si QD structures grown on Ge(111) substrates, the ESR signal with g-factor $g = 2.0022 \pm 0.0001$ and ESR line width $\Delta H \approx 1.2$ Oe was observed and attributed to the electrons localized in QDs. The g-factor value was explained by taking into account the energy band modification due to strain effects and quantum confinement. A strong Ge-Si intermixing in QD structures grown on Ge(001) is assumed to be main reason of unobserved ESR signal from QDs. The transport behavior confirms the efficient electron localization in Si QDs.

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INTRODUCTION

The long intrinsic electron spin coherence times in Si [1, 2] make Si-based heterostructures a natural choice for future quantum computation. Especially, an extremely long spin lifetime is expected in zero-dimensional structures, quantum dots (QDs), due to strong electron confinement in all three dimensions [3, 4]. One of the most promising Si-based systems with QDs is Ge/Si heterostructures, in which electrons are always localized in Si. In Ge/Si system with self-assembled Ge QDs the band offset makes QDs the potential barriers for electrons and the electron localization occurs in Si vicinity of Ge QDs due to the strain [5]. Strain-induced localization is not so strong and for its enhancement the multi-layered QD structures were proposed and realized [6]. In inverse system with Si QDs embedded in Ge matrix the potential wells for electrons are formed inside QDs and effective electron localization can be realized even in a one-layered Si QD structure. However, a small amount of Ge in Si QDs resulting from GeSi intermixing can frustrate all expectations of researchers [7] leading to shorter spin relaxation times due to a large spin-orbit interaction in Ge.

Different research groups tried to grow Si QDs on Ge [8–11] and faced the problem of Ge and Si intermixing caused by strong Ge segregation during Si growth on Ge(001) [12]. Moreover, in a Si-grown-on-Ge layer, dislocations are introduced much earlier [8, 10] than in a Ge-on-Si case, forced to decrease the thickness of deposited layer below the critical one for island nucleation. Our previous work [13] demonstrates a fundamental difference in the QD growth observed for Ge(111) and Ge(001) substrates. Si growth on Ge(001) is accompanied by strong Ge/Si intermixing even at sufficiently low (400 – 480°C) temperatures that leads to formation of two-dimensional

(2D) $\text{Ge}_x\text{Si}_{1-x}$ layer. The transition to three-dimensional (3D) growth is observed after deposition of 7 Si monolayers (MLs); 9-10 MLs are necessary to obtain the QD array with density of about 10^{11} cm^{-2} . The average Si content in these layers is $\approx 30\%$. On Ge(111) the Ge/Si intermixing is strongly suppressed due to a smaller Ge segregation during Si deposition. As a result, 3D growth of QDs with high Si content ($\approx 88\%$) [14] is observed without the underlying 2D layer formation (Volmer-Weber growth). Such high average Si content in QDs grown on Ge(111) suggests that this type of QDs can be considered as promising basic elements for future spintronic devices, provided that electrons are strongly localized in QDs.

This work is devoted to a study of electron localization in Si QDs grown on Ge(111) by transport and electron spin resonance (ESR) measurements. Conductance measurements demonstrate that transport is dominated by hopping via localized QD states. The ESR study reveals the ESR signal that can be attributed to QD electrons.

The paper begins with a description of experimental structures in section II. The theoretical consideration of electron states in QDs, including the estimation of spin relaxation time and electron g-factor, is presented in section III. Section IV demonstrates electron localization by means the transport measurements. The results of ESR study are given in section V.

EXPERIMENTAL DETAILS

Samples were grown by molecular beam epitaxy on Ge(111) substrates. Si QDs are formed by 2.5 Si bilayers (BLs) deposition at 400°C. The details of QD formation are described elsewhere [13]. The density of QD array obtained by STM is about $\sim 10^{11} \text{ cm}^{-2}$, the islands have a pyramidal shape with a hexagonal base, the average

lateral size l is about of 15 nm and aspect ratio $h/l \sim 0.1$. Raman measurements give the Si content $\approx 88\%$. Cross-sectional transmission electron microscopy (TEM) displays the dislocation-free structures.

For the transport measurements, the single-layered QD structures were grown on the Sb-doped Ge buffer layers with variation of Sb concentration in the range of $\sim 1 - 5 \times 10^{16} \text{ cm}^{-3}$. The thickness of Ge cap layer is 40 nm. The Al metal source and drain electrodes were deposited on the top of the structure and heated at 350°C in the Ar atmosphere to form reproducible Ohmic contacts. Since the ionization energy of Sb impurities in Ge is approximately 10 meV and the energies of the electron levels in Si QDs of this size are sufficiently larger, at low temperatures electrons should leave the impurities and fill the levels in QDs. The resistance along QD layer was measured by the four-terminal method. The temperature stability was controlled using a Ge thermometer.

To increase the response from the sample in ESR experiments, the structures with five Si QD layers separated by 10 nm Ge spacers were grown. The structures were capped with a $0.1 \mu\text{m}$ epitaxial n-Si layer (Sb concentration $\sim 5 \times 10^{16} \text{ cm}^{-3}$). The similar structures were grown on Ge(001) to verify the possibility of receiving ESR signal from Si QDs formed on the (001) substrates. The optimal conditions for QD formation on this type substrates were used, QDs were grown at 400°C by deposition of 10 MLs of Si. The ESR measurements were performed with a Bruker Eleksys 580 X-band ESR spectrometer using a dielectric Bruker ER-4118 X-MD-5 cavity. The sample was glued on a quartz holder, then the entire cavity with the sample was maintained at a low temperature in a helium flow cryostat (Oxford CF935).

ELECTRON SPIN STATES IN A SI QD

The effective mass calculations of the energy spectrum, including strain effects for the electron localized in a Si QD grown on Ge(111), were performed using the NEXTNANO program [15]. Based on the STM study [13], the Si truncated cone with base length $l=15$ nm and height $h=1.5$ nm was used as a Si QD model. The Si content was chosen to be 88% according to the Raman data. For comparison the energy levels for electron localized in a pure Si QD were calculated. The results of calculations show that ground state energy E_{QD} for the electron localized inside a pure Si QD is 123 meV, while, for $\text{Ge}_{0.12}\text{Si}_{0.88}$ QD, $E_{QD}=139$ meV (energy is measured from conduction band edge E_c in the Si QD layer). The energy of the first excited state in the pure Si QD is 146 meV (161 meV in the $\text{Ge}_{0.12}\text{Si}_{0.88}$ QD).

The obtained values of confinement energy are not so large as it is expected from the conduction band offset in GeSi heterostructures $\Delta U_{SiGe} \approx 430$ meV (derived from the data of Refs [16], [17]). This is a consequence of strain

effect on the band alignment for (111) orientation. The strain in the Si layer is close to the uniaxial compression along (111) direction and, opposite to the (001) case, does not induce the splitting of Δ -valley. The main strain effect results in the shift of the conduction band edge and corresponding decrease of the initial band offset down to $\Delta U_{SiGe} \approx 240$ meV.

The results of calculations can be used for the estimation of phonon-assisted spin relaxation time for an electron in the Si QD ground state. The spin reversal probability can be evaluated using the perturbation theory (PT). The matrix element of spin-phonon interaction $\langle \uparrow | \hat{H}_{ph} | \downarrow \rangle$ vanishes in the first order of PT [18]. The second order contribution is inversely proportional to the energy gaps between the ground and excited states. The calculated energy level spacing ≈ 20 meV is one order larger than that for gated QDs [19] allowing to expect a sufficient increase of the spin relaxation time. The spin relaxation time calculated in the same way as in our previous papers [20–22], turns out to be ~ 50 seconds at temperature 5 K and external magnetic field $H = 3455$ Oe. Such long spin relaxation time is a consequence of both small spin-orbit interaction in Si and strong electrons confinement in Si QDs.

With a knowledge of the energy spectrum and strain in the Si QD, it is possible to predict the electron g -factor value. For electrons in silicon, the g -factor can be evaluated through PT by the following equation [23]:

$$g = 2\mathbf{I} + \frac{2}{im} \Sigma_{\mu\nu} \frac{1}{E_{0\mu} E_{0\nu}} \{ \mathbf{h}_{0\mu} \mathbf{p}_{\mu\nu} \times \mathbf{p}_{\nu 0} + \mathbf{h}_{\nu 0} \mathbf{p}_{0\mu} \times \mathbf{p}_{\mu\nu} + \mathbf{h}_{\mu\nu} \mathbf{p}_{0\mu} \times \mathbf{p}_{\nu 0} \}, \quad (1)$$

where \mathbf{I} is the unit dyadic, $E_{0\mu} = E_0 - E_\mu$ is the energy gap between electron energy level E_0 and energy band E_μ contributing to g -factor correction, $p_{\mu\nu}$ is the momentum matrix element, and the spin-orbit interaction has the form of $\mathcal{H}_{so} = 2\mathbf{s} \cdot \mathbf{h}$.

Confinement and strain effects in the Si QD system lead mainly to the energy gaps modification, while the matrix elements of the momentum and spin-orbit interaction change negligibly. Then, for the estimation of the g -factor correction, one needs to know the positions of the main energy bands and the energy of the electron localized in a Si QD.

Electron energy E_0 can be written as $E_c + E_{QD}$, where E_c is the conduction band edge and E_{QD} is the energy of the electron ground state. Then g -factor correction acquires the form:

$$\delta g \sim \sum_{\mu\nu} \frac{C_{\mu\nu}}{(E_c + E_{QD} - E_\mu)(E_c + E_{QD} - E_\nu)}, \quad (2)$$

where $C_{\mu\nu}$ is the combination of matrix elements in curly brackets in Eq. 1. This allows us to separate the strain-induced shift of band edges E_c and $E_{\mu,\nu}$ and

the confinement-induced change of electron energy level E_{QD} .

The g -factor value in Si is mainly determined by the contribution of Δ_5 valence band and $2p$ core states [24]. For estimation, the shift of these band states can be taken roughly equal to the shift of the average energy of three uppermost valence bands E_{av} (bands of heavy, light and split-off holes). The NEXTNANO program allows the calculation of not only the electron energy in a Si QD, but the main energy bands position. The obtained E_c and E_{av} values are presented in Table 1 for a pure Si QD, for $\text{Ge}_{0.12}\text{Si}_{0.88}$ QD and for a bulk Si. The strain effect results in the upward shifts of E_c and E_{av} in a Si QD relatively to their original values in a bulk Si, $\Delta E_c = 0.2$ eV and $\Delta E_{av} = 0.113$ eV, correspondingly. Since the electron energy is $E_{QD} = 0.123$ eV, then the energy gap $E_{0\mu,\nu}$ determining the correction to the g -factor increases by ~ 0.2 eV. Such large change of $E_{0\mu,\nu}$ allows us to expect a sufficient change of the electron g -factor value in Si QDs.

The smallness of E_{QD} , as compared to the distance between the E_c and the nearest valence band at the Δ -point ($E_c - E_{\Delta_5} = 4.27$ eV) [25], makes it possible to use the Taylor series expansion of expression δg in parameter E_{QD} for estimation of g -factor value. As a result, we obtain linear dependence $\delta g \sim K \cdot E_{QD}$, with the coefficient K being determined by the magnitude of the energy gaps and matrix elements of momentum and spin-orbit interaction. Similar dependence of g -factor value on the electron binding energy E_b for As, P and Sb impurities in Si was plotted in the paper by Young *et al.* [26] using the empirical data measured by Feher [7]. The extrapolation of this linear dependence allows the authors [26] to predict the g -factor value for electrons on conduction band edge $g_c = 1.9995$. High-precision ESR measurements have shown an excellent agreement between the measured and expected g_c value. We suppose that the empirical linear dependence observed in the work [26] is also the result of Taylor expansion of δg , but in the small parameter E_b . Then the slope of this line gives the coefficient K for unstrained bulk Si. In the strained Si this coefficient is changed due to the energy bands shift and, in the simple case of two band approximation, it takes the value of $K' \sim C/(E'_c - E'_{\Delta_5})^3$, where E'_c and E'_{Δ_5} are strain-modified energy bands positions. It is possible to estimate K' using the energy bands modification from Table 1 and coefficient K from the empirical dependence given in the paper [26].

The black solid line in Fig. 1 represents the extrapolation of the linear dependence of g -factor on the electron binding energy for impurities (filled squares) in bulk unstrained Si [7]. The red dash line demonstrates the modification of this linear dependence for the strained Si QD layer in the simple case when g -factor is determined by the contribution of the only one nearest energy band. The lines are separated along the energy axis, because

TABLE I: The main energy bands positions and ground state energy values of the electron in a Si QD calculated using the NEXTNANO program in the effective mass approximation taking into account strain effects. The reference point for electron energy was chosen to be the same as in the work [27]

energy band	pure Si QD	$\text{Ge}_{0.12}\text{Si}_{0.88}$ QD	Si bulk
E_c	2.47	2.45	2.27
E_{av}	1.203	1.246	1.09
$E_{QD} + E_c$	2.593	2.589	-

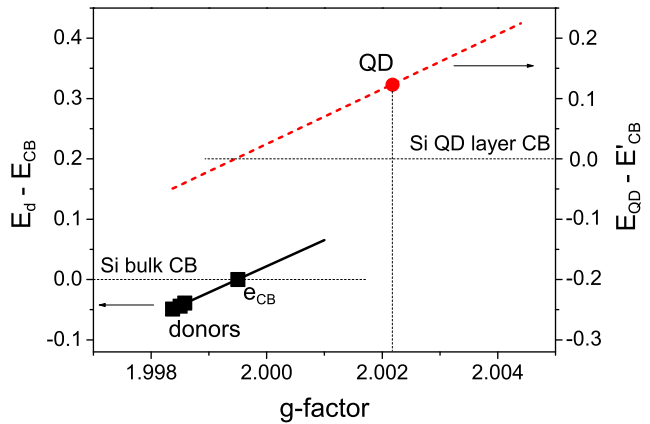


FIG. 1: An extrapolation of the g -factor linear dependence on the electron binding energy (black solid line) for the conduction-band electrons (CB) and three shallow donors in bulk Si (filled squares). The red dashed line is the modification of this linear dependence for the case of a strained Si QD layer. The red circle corresponds to the g -factor value for a pure Si QD.

the conduction band edge in the case of the Si QD layer is shifted to the higher energies due to the strain. The red dashed line has a slope slightly different from the slope of the line for the donors in unstrained bulk Si. The g -factor dependence is presented for a pure Si QD because the difference between the slope of the lines plotted for QDs with 100% and 88% Si content is negligible. The difference will be observed if one takes into account the change of the constant C due to Ge admixing. For the electrons localized in a 100% Si QD, the linear dependence allows us to predict $g_{QD} = 2.00218$ (red circle in Fig. 1).

ELECTRON LOCALIZATION: TRANSPORT PROPERTIES

Transport measurements in disordered systems allow one to understand whether the carriers are strongly localized or not. In the case of strong localization, the transport is carried out by hopping between localized states [28]. In the opposite case of delocalized carriers, the transport is described by the classical Drude the-

ory with quantum corrections to the conductivity derived from the weak localization theory [29]. Different transport mechanisms manifest themselves in different temperature dependencies of the conductance and different magnetoresistance behaviors.

It was previously shown [30] that the lateral transport in two-dimensional Ge QD array with QD sizes and density similar to the Si QD system under study is described by variable range hopping via strong localized QD states taking into account long-range Coulomb interaction (Efros-Shklovskii (ES) law [31]). To confirm the strong electron localization in Si QDs and check whether the mechanism of electron transport in Si QD samples is similar to the hole transfer in Ge QD samples, the temperature dependence of conductance, magnetoresistance (MR) and photoconductance were studied. The temperature dependencies of the lateral conductance for Si QD samples with two different doping levels are demonstrated in Fig. 2. The high temperature part of the curves corresponds to the ionization of impurities to the conduction band. Analysis of the low temperature parts (<10 K) shows that the temperature dependencies of conductance for both samples are described by ES law confirmed the strong carriers localization in the structure under study. The small temperature of the transition from the band to hopping transport, as compared to Ge/Si QDs structures [30], is explained by smaller ionization energy of Sb in Ge than in Si.

An additional heating at $470 - 600^\circ\text{C}$ does not change the conductance of the system. Previously, in the Ge/Si heterostructure with Ge QDs grown on Si(001), annealing at $550 - 625^\circ\text{C}$ temperatures resulted in a considerable increase of the conductance up to the observation of transition from hopping to the diffusion regime [30]. This was explained by smoothing of the localization potential and the corresponding increase of the wave functions overlapping due to a smearing of the QDs caused by Ge-Si intermixing. In the structures under study the diffusive smearing of Si QDs is suppressed due to a larger stability of Ge/Si interfaces with (111) orientation than that with (001) [32].

The photoconductance kinetics of sample 2 under illumination with the $1.5 \mu\text{m}$ wavelength light is shown in the inset to Fig. 2. Slow kinetics under switch on and switch off the illumination, accompanied with the persistent photoconductance effect, was already observed for Ge QD structures and explained by the spatial electrons and holes separation in type-II QDs [33]. We suppose that the same mechanism is responsible for the observed photoconductance behavior in the Si QD system.

The MR data of Si QD sample 1 measured in the perpendicular magnetic field at 2 K and 4.2 K are demonstrated in Fig. 3. One can see that MR is negative in weak magnetic fields, then it crosses to a positive MR with an increase of the magnetic field. Recently, we have observed such MR behavior in the Ge QD structures

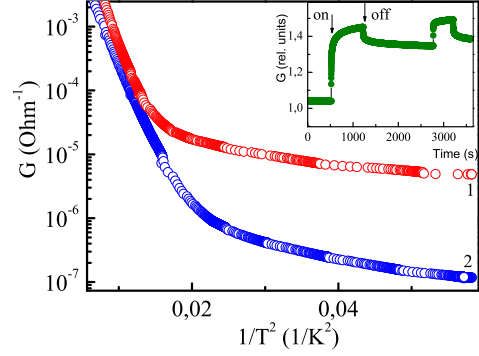


FIG. 2: Temperature dependencies of the conductance for QD structures with different doping levels. The photoconductance kinetics of sample 2 under illumination with the $1.5 \mu\text{m}$ wavelength is shown in the inset.

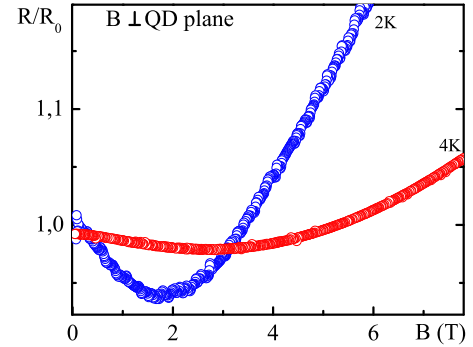


FIG. 3: Magnetoresistance of Si QD structure in perpendicular magnetic field at different temperatures.

grown on Si [34]. The positive MR in high magnetic fields was explained by suppression of the conductance due to shrinking of the carrier wave functions, whereas a negative weak-field MR was referred to the weak localization contributions to the conductance of clusters with closely located QDs.

Thus, obviously similar transport properties of Ge and Si QD systems with close structural parameters (QD size, shape and areal density) allows us to conclude that a strong localization of electrons occurs in Si QDs as well as a hole localization in Ge QDs.

ESR STUDY

The structures with Si QDs grown on Ge(111) were investigated using ESR spectroscopy at $T=4.5$ K. The observed ESR signal is shown in Fig. 4. The approximation of the experimental curve demonstrates that the ESR signal is a superposition of two ESR lines - one wide

ESR line with the width of $\Delta H \approx 6$ Oe and the second narrow ESR line with $\Delta H \approx 1.2$ Oe. Both signals have isotropic g -factor $g = 2.0022 \pm 0.0001$ in spite of their different origin. The wide ESR line has a Gaussian shape that indicates a non-homogeneous broadening of the signal. Heating at 650°C for 10 min leads to complete disappearance of this ESR line. A very similar signal was observed in the paper [35] for the oxidized Ge substrates. We attribute this signal to the defects in epitaxial Si/Ge layers or in the GeO_x layer. The test structure grown in the same conditions, but without QDs shows the only wide ESR signal.

The narrow ESR signal has a Lorentzian shape. Heating does not affect both ESR line width and g -factor value suggesting that the signal belongs to the electrons localized in Si QDs. The annealing stability of the g -factor indicates the negligible Ge/Si intermixing. From the invariance of the ESR line width it can be assumed that spin relaxation time T_2 remains the same (for Lorentzian lines $\Delta H \sim 1/T_2$). It is well known that the stronger the carrier confinement, the longer the spin relaxation time is [4]. It means that the electron localization radius does not change considerably after annealing that is in a good agreement with the results of conductance measurements.

The isotropy of the g -factor does not contradict the Si QD origin of narrow signal. The g -factor anisotropy obtained earlier for QDs grown on (001) substrates results from Δ -valley splitting induced by the uniaxial strain along (001) [36]. In the structures under study the uniaxial strain along (111) does not affect the conduction band minima at the Δ -point [37]. The observed isotropic g -factor is explained by missing the Δ -valley splitting for thin Si QD layers grown on the Ge (111) substrate.

The shape of narrow ESR line is asymmetric and close to the Dysonian one [38] that can be explained by the admixture of dispersion signal [39]. The appearance of noticeable dispersion contribution is the characteristic feature of the samples with non-zero conductivity. As shown in our previous work [40], the well pronounced asymmetry of ESR line shape can be resulted from the hopping transport across the QD array. However, there is one more possible reason of ESR line asymmetry. Earlier [7] for donor electrons in Si the asymmetry of ESR line was induced by small admixture of Ge atoms ($\sim 1\%$). In Si QD system the asymmetry of ESR line can arise due to penetration of electron wave function into Ge barrier. It should be noted that ESR lines from the defects in Si and Ge usually have symmetric shape and are characterized by a larger line width, especially for Ge, in which ESR line broadening reaches tens of oersteds.

The g -factor value $g_{\text{QD}} = 2.0022$ obtained in ESR experiments is in a good agreement with predicted in Sec. 3 value $g = 2.00218$ (red circle in Fig. 1) and is sufficiently larger than that observed earlier for the electrons in Si. In bulk Si, the g -factor of conduction electrons is

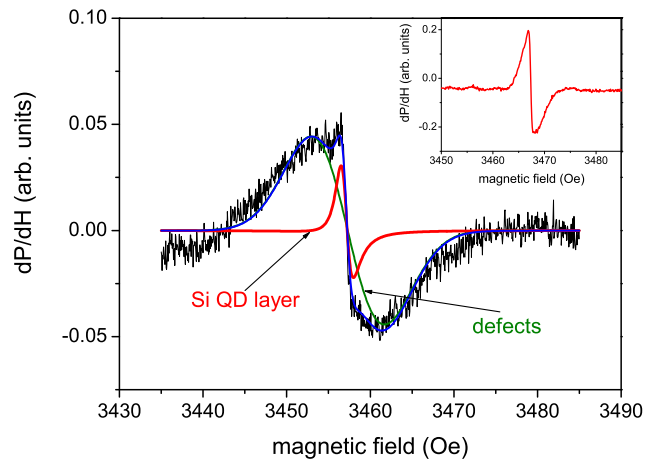


FIG. 4: The ESR signal obtained for the heterostructure with 5 layers of Si QDs; microwave power and frequency $P=0.063$ mW, $\nu = 9.68849$ GHz, temperature $T=4.5$ K. Solid lines are the approximation of the experimental signal. The ESR signal after annealing, $\nu = 9.71679$ GHz, is shown in the inset.

$g_{ce} = 1.9987$, while for different quantum-confined systems, g_{ce} varies from 1.9986 to 2.0007 [41, 42], depending on the microscopic structure of quantum wells. A larger value of electron g -factor in Si QDs is explained by strong electron confinement and strong modification of band structure due to strain.

The fact that the experimental g -factor value is close to the predicted one for pure Si QD suggests that QDs in the structures under study contain pure Si core. This should provide long spin relaxation times. However, the spin transverse time, determined from the ESR line width is about of $T_2 \sim 10^{-7}$ s. The longitudinal spin relaxation time T_1 can be estimated from microwave power dependence of the ESR signal. In our case the saturation of the ESR signal does not observed up to microwave power 0.4 mW that corresponds to the limitation of times $T_1 \leq 10\mu\text{s}$. Such short times can be related to spin relaxation induced by tunneling between QDs. Recently the Dyakonov-Perel mechanism was proposed for explanation of fast spin relaxation in dense QD arrays [36]. However, this mechanism should reveal itself in the anisotropy of the ESR line width that is not observed in the structure under study. The spin relaxation in Si QD system is more likely provoked by intervalley transitions [43] when electron passes the Si/Ge/Si heterointerfaces during tunneling between QDs. To increase the spin relaxation time, one needs to suppress the tunneling by spatial separation of QDs.

The ESR study of the Si/Ge structure with QDs grown on Ge(001) shows the only wide ESR signal that totally disappears after 650°C annealing. The narrow ESR signal from Si QDs grown on Ge(001) could not be resolved plainly because of excessive signal broadening due to a

significant admixture of Ge into Si during the QD growth. The GeSi intermixing is the result of Ge atoms segregation during Si deposition. By means this segregation the total energy of the system decreases, since Ge(001) surface has smaller surface energy than Si(001) one [11]. In contrast, the Ge segregation during Si growth on Ge(111) is strongly suppressed, because the difference between Ge and Si surface energies for (111)-oriented samples is much smaller [32], making it possible to detect the ESR signal from Si QDs grown on Ge(111).

CONCLUSION

In summary, the effective electron localization in Si QDs was confirmed by transport and ESR measurements. Temperature dependence of the conductance is well described within a variable range hopping model indicating a strong electron localization in the system. The photoconductance behavior is typical of the type-II QDs with one type of localization carriers. The high Si content in QDs grown on Ge(111) allows us to detect the narrow ESR signal from QD electrons. It was shown that the isotropic ESR signal with g -factor $g = 2.0022$ and line width $\Delta H \approx 1.2$ Oe is related to the electrons localized in QDs. An extrapolation of the empirical linear dependence of the g -factor on the binding energy of localized electrons in Si [26] was used to predict the g -factor value for electrons in Si QDs. The experimental value is in a good agreement with the g -factor estimated by taking into account the strain effects and strong electron confinement in Si QD.

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